# Hydrogenation of Nd-Fe-B magnet powder under a high pressure of hydrogen

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**Abstract**

The hydrogenation of $Nd_2Fe_{14}B$ under a high pressure of hydrogen has been investigated for the first time. At the heat-treatment temperature of 600°C, the almost complete decomposition of $Nd_2Fe_{14}B$ into $NdH_{2+x}$ and $\alpha$-Fe is observed, although a rather long heat-treatment time is necessary to achieve the sufficient hydrogenation. The desorption of hydrogen from $NdH_{2+x}$ does not occur in the furnace-cooling process.

*Keywords:* hydrogenation, Nd-Fe-B magnet, HDDR process

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## 1. Introduction

Toward the improved magnetic-properties of the Nd-Fe-B permanent magnet, one of important technologies is the hydrogenation of $Nd_2Fe_{14}B$, which is employed in the so-called HDDR (hydrogenation, disproportionation, desorption, recombination) process[1]. The HDDR-processed magnet possesses a submicron grain-size, contributing a higher coercivity. $H_2$ atmosphere with rather low pressure ($\sim$ 1 atm) is introduced in the hydrogenation process. After the heat-treatment at 750 $\sim$ 900 °C for 30 min $\sim$ 3 h, the magnet disproportionates into Nd hydride ($NdH_{2+x}$), $\alpha$-Fe and $Fe_2B$.

Several groups have reported the reactive milling of Nd-Fe-B powder under hydrogen at room temperature[2, 3, 4]. The mechanical activation of Nd-Fe-B powder leads to the decomposion into $NdH_{2+x}$ and $\alpha$-Fe, although we need high pressure of $H_2$ gas ($\sim$ 1 MPa). These studies motivated us to investigate the conventional hydrogenation of Nd-Fe-B under a high pressure of $H_2$ gas, which has not been reported. In this study, we have studied the hydrogenation of Nd-Fe-B under a high pressure of hydrogen.

## 2. Experimental method

We used a commercial Nd-Fe-B magnet. The ground magnet powder, after demagnetization, weighting approximately 0.45 g were placed in a home-made cell with the volume of about 7.5 cc. After evacuating the cell, we introduced $H_2$ gas of 0.45 MPa. The cell was heated to the temperature ranging from 500 °C to 600°C, taking 2 h, by an electric furnace, and kept at that temperature for 6 to 84 h, followed by a furnace-cooling. The heat-treated samples were evaluated using the powder X-ray diffraction (XRD)

pattern (Cu-K$\alpha$ radiation). We also hydrogenated Nd-Fe-B powder in a continuous $H_2$ gas flow at 600°C for 12 h, in which the heating and cooling rates are the same as those in the above-mentioned hydrogenation process.

## 3. Results and discussion

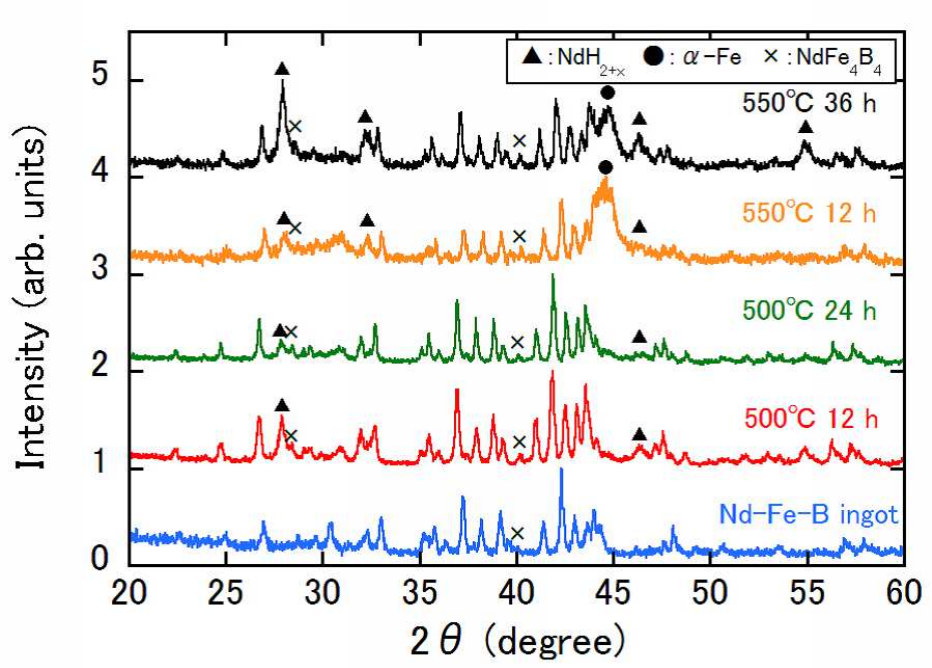


Figure 1: XRD patterns of samples after heat-treatment at 500 or 550°C . The XRD pattern of Nd-Fe-B ingot is also shown.

The XRD patterns of samples heat-treated at 500°C for 12 and 24 h are shown in Fig. 1, where the pattern of Nd-Fe-B ingot is also exhibited. The pattern of Nd-Fe-B ingot is mainly composed of $Nd_2Fe_{14}B$ in addition to the minor phase of $NdFe_4B_4$. The peaks of $Nd_2Fe_{14}B$ in the samples heat-treated at 500°C are shifted to lower $2\theta$ values compared to those of the

starting ingot. This implies the lattice expansion of $Nd_2Fe_{14}B$ due to the insertion of hydrogen[5]. The decomposition of $Nd_2Fe_{14}B$ does not occur because of no appearance of $\alpha$-Fe, which is confirmed by further extending the heat-treatment time to 84 h. The $NdH_{2+x}$ phase denoted by triangles would be generated from a Nd-rich phase[6].

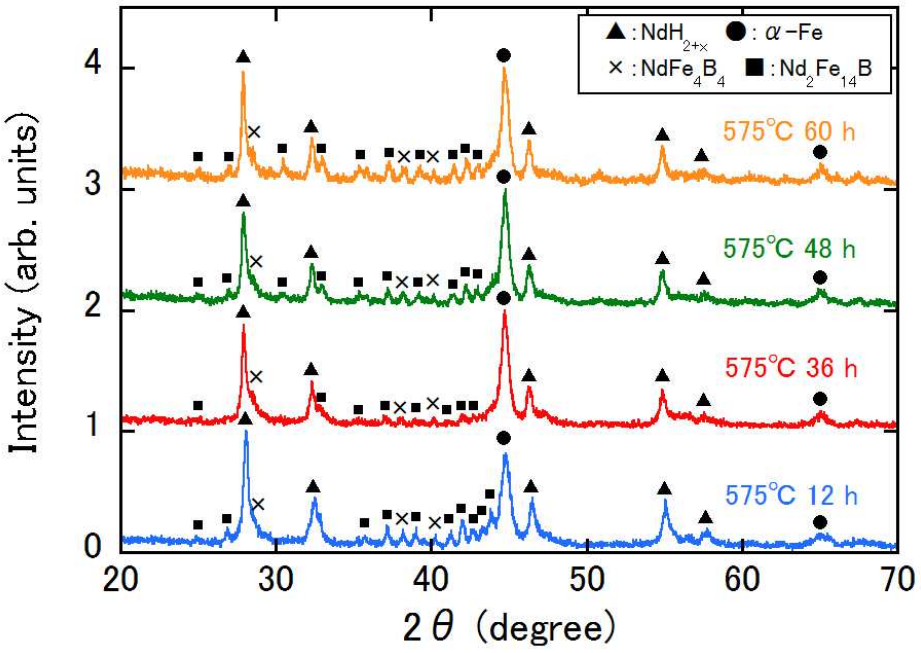


Figure 2: XRD patterns of samples after heat-treatment at 575°C .

Fig. 1 also shows the XRD patterns of samples heat-treated at 550°C for 12 and 36 h. The shift of peaks of $Nd_2Fe_{14}B$ does not occur in each sample, indicating no insertion of hydrogen in $Nd_2Fe_{14}B$. Although $Nd_2Fe_{14}B$ is not completely decomposed, the simultaneous appearance of $NdH_{2+x}$ and $\alpha$-Fe means the partial hydrogenation of $Nd_2Fe_{14}B$.

As shown in Fig. 2, the hydrogenation of $Nd_2Fe_{14}B$ proceeds by increasing the heat-treatment temperature to 575°C. The peaks of $NdH_{2+x}$ and $\alpha$-Fe phases dominate over those of unreacted $Nd_2Fe_{14}B$ and $NdFe_4B_4$, which is observed throughout the heat-treatment time of interest.

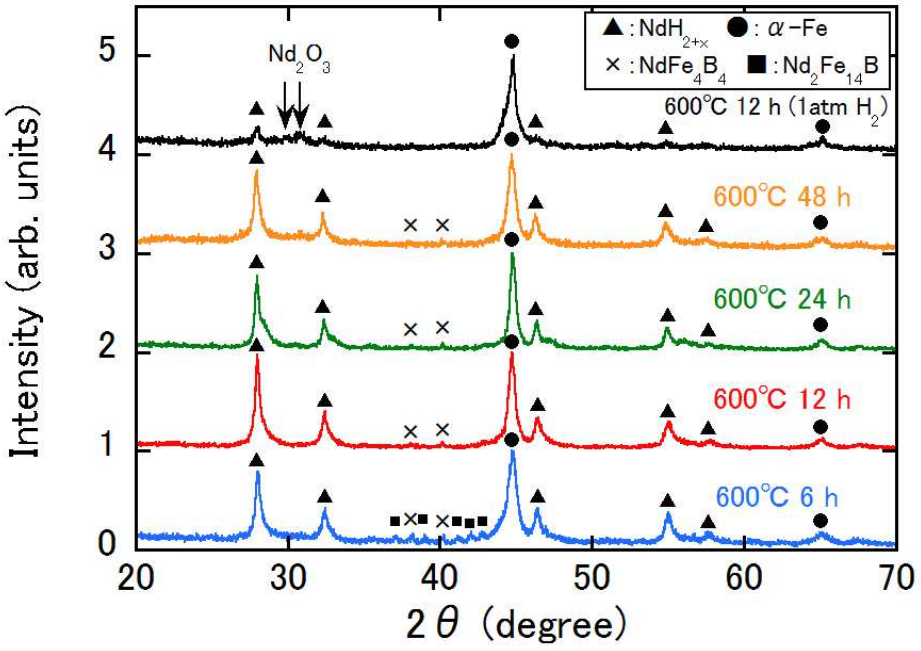


Figure 3: XRD patterns of samples after heat-treatment at 600°C .

At the heat-treatment temperature of 600°C for longer than 12 h, the almost complete decomposition of $Nd_2Fe_{14}B$ is observed (see Fig. 3). We cannot say whether the $Fe_2B$ phase, which is detected after the disproportionation in the HDDR process, exists or not. To investigate the effect of high-pressure $H_2$ gas, we have carried out the hydrogenation of Nd-Fe-B in a continuous $H_2$ gas flow. The XRD pattern of the sample is shown at the

top of Fig. 3. We have found that, even at the heat-treatment temperature lower than that in typical HDDR process, the complete decomposition of $Nd_2Fe_{14}B$ into $NdH_{2+x}$ and $\alpha$-Fe can be achieved for the prolonged heat-treatment time. However the peak intensity of $NdH_{2+x}$ is very weak compared to that in samples hydrogenated under high $H_2$-pressure. In addition, $Nd_2O_3$ phase is detected (see arrows in Fig. 3). During the furnace-cooling, some hydrogen might desorb from $NdH_{2+x}$ transforming into $Nd_2O_3$. On the other hand, the samples hydrogenated under high $H_2$-pressure do not show the desorption of hydrogen after the furnace-cooling.

## 4. Summary

We have investigated the hydrogenation of Nd-Fe-B powder under the high pressure of hydrogen. The hydrogenation of $Nd_2Fe_{14}B$ proceeds almost completely at the heat-treatment temperature of 600°C, although the prolonged heat-treatment time is necessary for the sufficient decomposition of $Nd_2Fe_{14}B$.

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